

## Synthesis and Characterization of Ag/TiO<sub>2</sub> Nanoparticles using *Mirabilis jalapa* Plant Extract

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### Abstract

Green synthesized nanomaterials have been widely developed because of their less toxicity, low energy process, environmentally friendly, effective, cheap, and pollution-free. Green synthesis of silver doped titanium dioxide nanoparticles (Ag/TiO<sub>2</sub> NPs) was carried out using *Mirabilis jalapa* plant extract. The plant extract was used as a reducing agent. The functional groups, morphology, and crystalline structure of as-synthesized Ag/TiO<sub>2</sub> NPs were investigated by FT-IR, FESEM, and XRD. Analysis by FESEM confirmed that the morphology of Ag/TiO<sub>2</sub> NPs is spherical with an average size of ~ 400 nm. Crystallite size for the Ag/TiO<sub>2</sub> NPs was calculated by the Scherrer formula and the average size found to be in the range of 15.72 nm. The result of XRD analysis showing the fcc structure for metallic silver and TiO<sub>2</sub> particles in the anatase and rutile phases.

**Keywords:** Green synthesis, nanoparticles Ag/TiO<sub>2</sub>, *Mirabilis jalapa*

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### 1. INTRODUCTION

Nanoparticles (NPs) have been widely developed because of their unique properties compared to their bulk material. Nanoparticles have unique characteristics due to the size, morphology, shape, and distribution (Amaliyah et al., 2020). Nanoparticles with green synthesis methods have been developed in the last decade. Green synthesis is a facile synthesis method that has the advantage of being an approach environmentally friendly, less toxic, low energy process, effective, inexpensive, and pollution-free. Plants, microorganisms, algae, or extracts plants as reducing agents can be used to control the shape and size of nanoparticles (Jegadeeswaran et al., 2016). The green synthesis nanoparticles show good morphology and stability (Rao et al., 2019).

Titanium dioxide, zinc oxide, silver, gold, copper, graphene oxide nanoparticles have been synthesized by green chemistry and an environmentally friendly approach (Jegadeeswaran et al., 2016). Titanium dioxide (TiO<sub>2</sub>) nanoparticles with unique surface

chemistry, morphologies, good stability, non-toxicity, and photocatalytic oxidation ability have been used in several applications. To increase the photocatalytic activity of TiO<sub>2</sub>, various modifications have been made with noble metals (Au, Ag) (Liu et al., 2019). The energy bandgap of TiO<sub>2</sub> NPs manipulated by a doped of metals. Ag/TiO<sub>2</sub> NPs have been synthesized due to non-toxic, high availability, and low-cost material (Rao et al., 2019).

Recently, Ag/TiO<sub>2</sub> NPs have been synthesized from various plants including *Padina tetrastrum* (seaweed), *Acacia nilotica*, *Euphorbia heterophylla* (Atarod et al., 2016; Jegadeeswaran et al., 2016; Rao et al., 2019). *Padina tetrastrum* (seaweed) plant extract has been successfully used in the synthesis of Ag/TiO<sub>2</sub> nanocomposites. Seaweed extract acted as a capping agent and reducing agent in the formation of nanocomposites. The addition of plant extracts controlled the shape and size of the nanocomposites. The particle size of Ag/TiO<sub>2</sub> nanocomposite synthesized with seaweed was 25 nm (Jegadeeswaran et al., 2016).

However, the green synthesis of Ag/TiO<sub>2</sub> NPs using plant extracts as a reducing agent is still rarely investigated. Phytochemical tests of *Mirabilis jalapa* shown that the plant contains flavonoids, saponins, tannins, and terpenoids (Oktaviana, 2018). These compounds have the potential as a capping agent and reducing agent for the synthesis of NPs. *Mirabilis jalapa* has been used for the synthesis of Ag/ZnO nanoparticles. This plant is used as a reducing agent for the bioreduction of zinc acetate and AgNO<sub>3</sub> in the formation of Ag/ZnO nanoparticles (Sumbal et al., 2019). In this study, we reported *Mirabilis jalapa* plant extract which can be used to synthesizing Ag/TiO<sub>2</sub> NPs. Ag/TiO<sub>2</sub> NPs were characterized using UV-Vis, FTIR, XRD, and FESEM.

## 2. MATERIALS AND METHODS

### Material

The materials used in this study were titanium (IV) isopropoxide (TTIP) (Sigma Aldrich 98%), silver nitrate (AgNO<sub>3</sub>) (Merck), ethanol, and *Mirabilis jalapa* plant extract.

### Procedure

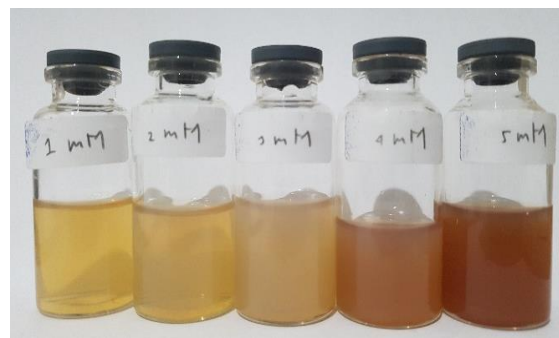
In the Ag/TiO<sub>2</sub> synthesis, the Ag nanoparticles were synthesized separately first. A total of 50 ml of extract from *Mirabilis jalapa* was added to a 0.003 M AgNO<sub>3</sub> solution. Then the solution was stirred at 100 °C until the color changed. The solution formed is diluted using aquadest. The Ag/TiO<sub>2</sub> synthesis was carried out by adding 50 ml of ethanol to 5 ml of titanium (IV) isopropoxide solution and stirred for 1 hour. The solution was added with AgNO<sub>3</sub> solution and *Mirabilis Jalapa* extract solution and continued by stirring for 1 hour. The solution was calcined at 450 °C for 2 hours. The characterization of the material was carried out using UV-Vis, FT-IR, XRD and SEM.

## 3. RESULTS AND DISCUSSION

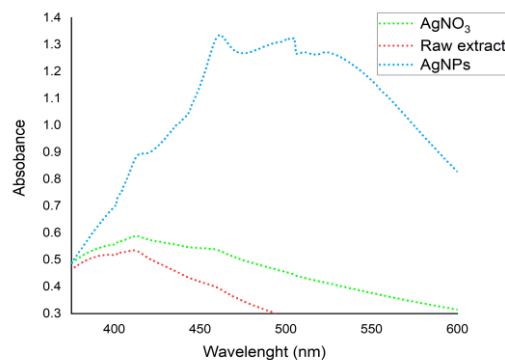
In this study, Ag/TiO<sub>2</sub> nanoparticles were synthesized using a solution of *Mirabilis jalapa* plant extract as a solvent. Green synthesis of Ag/TiO<sub>2</sub> using *Mirabilis jalapa* plant extract has been investigated which is a facile, cost-effective, non-toxic, environmentally friendly, and efficient method for utilization of *Mirabilis jalapa* plant extract. The plant extract acted both as a reducing

agent and stabilizer in the synthesis of nanoparticles.

The addition of *Mirabilis jalapa* plant in the aqueous extract to AgNO<sub>3</sub> solution generated a color change from colorless to dark brown after stirring 1 h of reaction. The color changes indicated that AgNPs have been successfully formed (Sumbal et al., 2019). The color changes depending on the concentration of AgNO<sub>3</sub>. The synthesis of AgNPs with various concentration variations can be seen in **Figure 1**. AgNPs were synthesized by AgNO<sub>3</sub> 5 mM which showed the optimum concentration of AgNO<sub>3</sub>.



**Figure 1.** AgNPs solution with various concentrations of AgNO<sub>3</sub> in aqueous solvent.



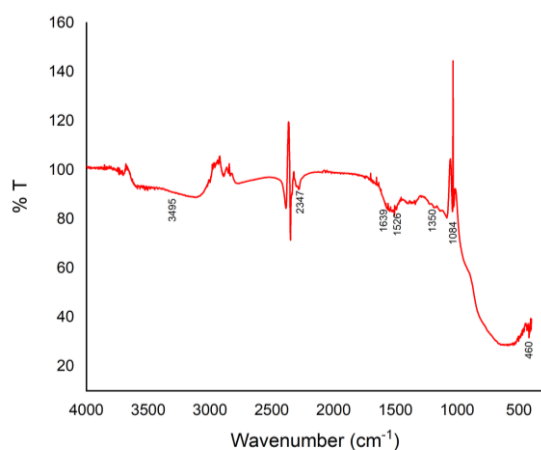
**Figure 2.** UV-Vis spectra of AgNPs with SPR band at 460 nm

Figure 2 showed that a sharp absorption band at 460 nm indicates the particles had dispersed. There was a shift in wavelength due to the addition of *Mirabilis Jalapa* extract which indicated that there was oxidation due to the addition of *Mirabilis Jalapa* (Fatimah & Mutiara, 2016). The surface plasmon resonance (SPR) band of AgNPs was exhibited at 390 – 500 nm which showed the characteristic of the absorption band of silver nanoparticles. Round-shaped nanoparticles

were confirmed at 300 to 700 nm which indicated a single surface plasmon resonance (Yousaf et al., 2020). The broadening of the absorbance affected the shape and size of the NPs (Gopalakrishnan *et al.*, 2019).

### Characterization of *Mirabilis jalapa* mediated Ag/TiO<sub>2</sub> nanoparticles

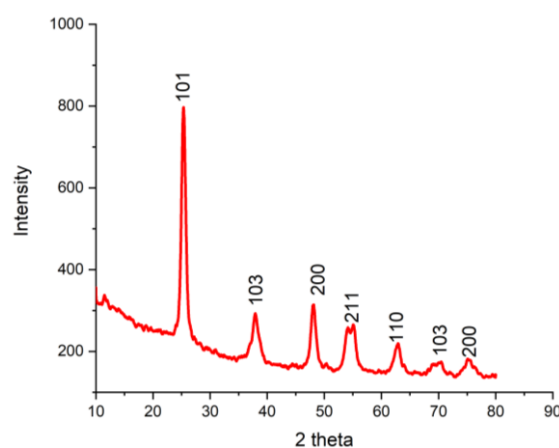
The FTIR analysis of Ag/TiO<sub>2</sub> nanoparticles shown in **Figure 3**. FTIR analysis is to determine the functional groups present in the synthesized Ag/TiO<sub>2</sub> nanoparticles. The peak at 1350 cm<sup>-1</sup> is characteristic of the Ti–ligand bond (Alsharaeh et al., 2017; Jegadeeswaran et al., 2016). The peaks shown at 1639 and 3495 cm<sup>-1</sup> indicated the presence of –OH bending and –OH stretching, respectively. The band located at 460 cm<sup>-1</sup> was generated due to the oxide structure formation (Jegadeeswaran *et al.*, 2016). Furthermore, the peak at 2347 cm<sup>-1</sup> indicated the transition metal carbonyls group (Rao *et al.*, 2019). The peaks at 1084 cm<sup>-1</sup> and 1526 cm<sup>-1</sup> represent the C=C aromatic ring and C – OH stretching vibration, respectively. The presence of the functional groups from polyphenolics indicated the phenolics compound in the plant extract. The reduction of metal ions and formation of nanocomposite could be probably due to the presence of a phenolic compound (Atarod *et al.*, 2016).



**Figure 3.** FTIR analysis of green synthesized Ag/TiO<sub>2</sub> nanoparticles.

Figure 4 shows the XRD pattern of Ag/TiO<sub>2</sub> NPs. The result showed that there were eleven peaks of the diffractogram, consisting four peaks of titanium substrate, five peaks of anatase TiO<sub>2</sub> crystals and 2 peaks

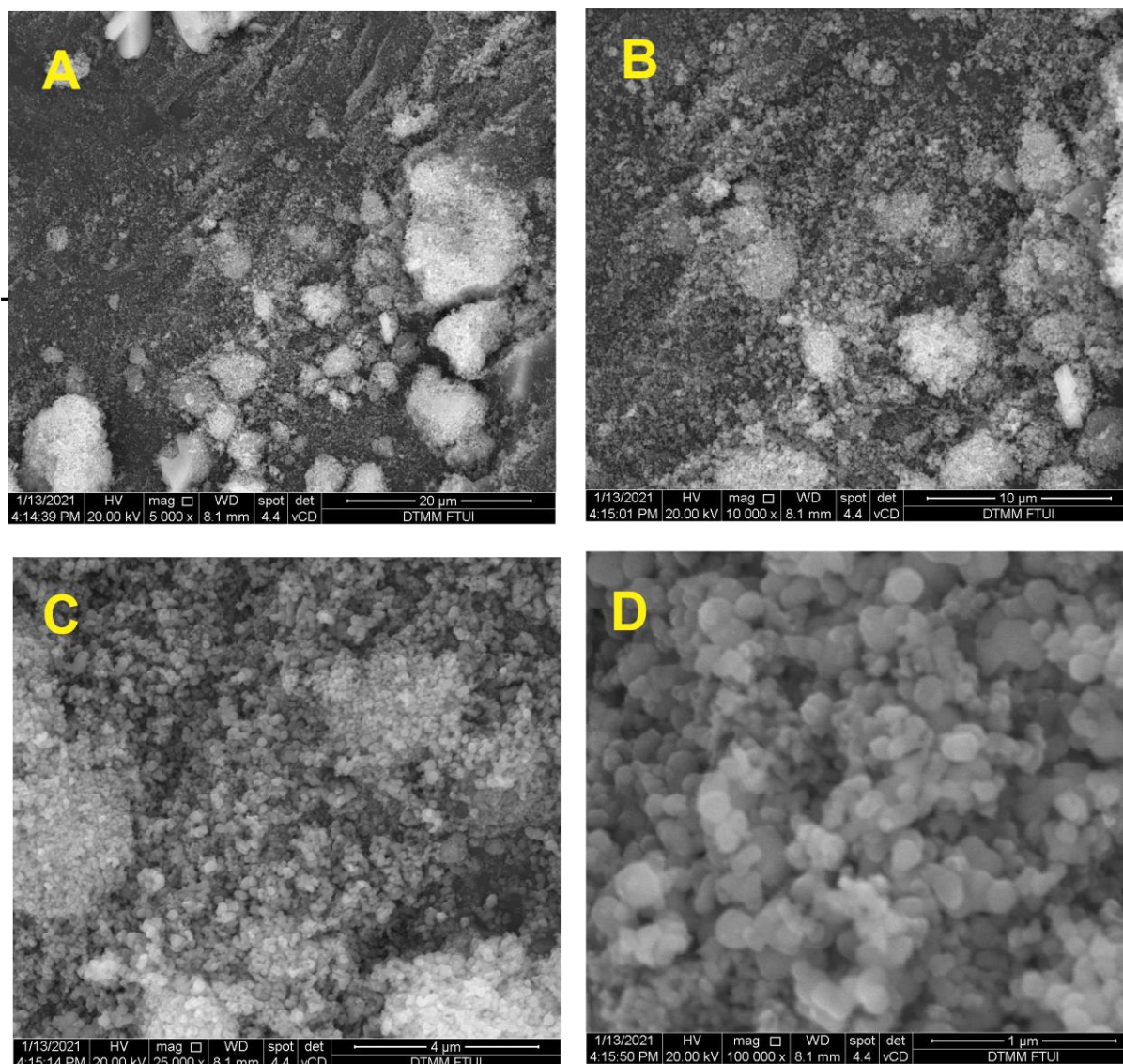
of Ag crystals. The peaks in the synthesized Ag/TiO<sub>2</sub> nanoparticles were compared with the JCPDS data No. 1272 for TiO<sub>2</sub> anatase ; 1294 for TiO<sub>2</sub> rutile and 040783 for Ag. All peaks at 23.374°, 37.008°, 48.068° and 55.000° corresponds to the crystal planes of (101), (103), (200) and (211) respectively, indicating the formation of TiO<sub>2</sub> NPs in anatase phase. The peak at 62.743°, 70.457°, 74.276° and 76.175° corresponds and marked by their indices crystal planes of (110), (103), (200) and (112) respectively, which proves the formation of TiO<sub>2</sub> NPs in rutile phase had successfully synthesized (Purnomo et al., 2018). The peak of 37.994° and 45.000° respectively, indicating the present of Ag. Crystallite size for the Ag/TiO<sub>2</sub> nanoparticles was calculated by the Debye-Scherrer equation and the average size was found to be around 15.72 nm.



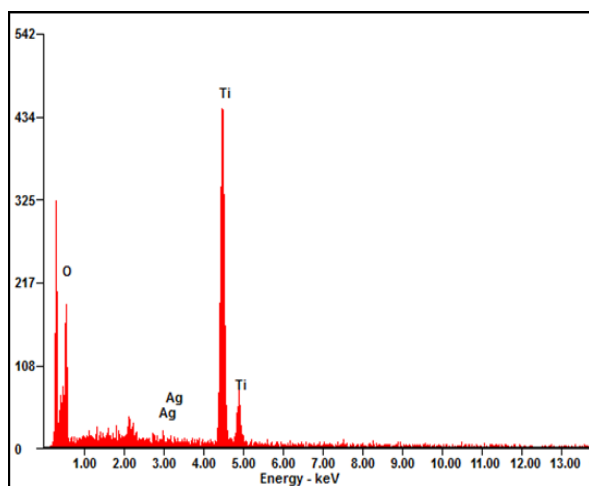
**Figure 4.** XRD analysis of green synthesized Ag/TiO<sub>2</sub> nanoparticles.

To determine the morphology and size of Ag/TiO<sub>2</sub> nanoparticles, characterization was carried out by using FESEM. The result of the FESEM analysis is shown in **Fig. 5**. The average size of the Ag/TiO<sub>2</sub> nanoparticles was  $\pm 160$  nm according to the ImageJ software. The morphology of the synthesized Ag/TiO<sub>2</sub> nanoparticles is spherical-shaped. This finding similar to the results reported by Sumbal et al., 2019 and Yousaf et al., 2020 that the Ag nanoparticles with extract plant in aqueous solution produce a spherical shape. The morphology variation of nanoparticles depend on the type of metal, conditions of the reaction, concentration of salt, and the composition of plant extracts (Sumbal *et al.*, 2019).



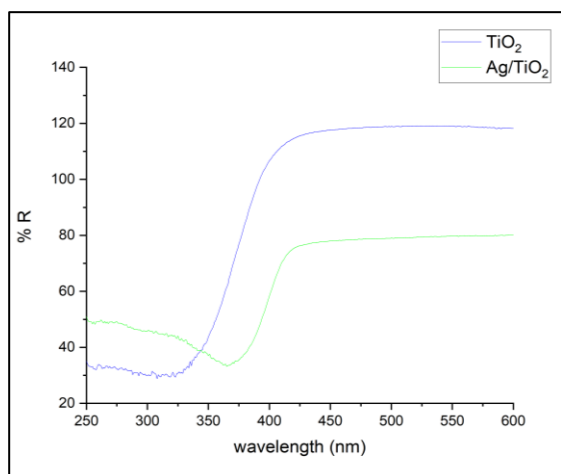


**Fig. 5.** FESEM analysis of green synthesis Ag/TiO<sub>2</sub> nanoparticles a) 5.000 magnification; b) 10.000 magnification; c) 25.000 magnification and d) 100.000 magnification

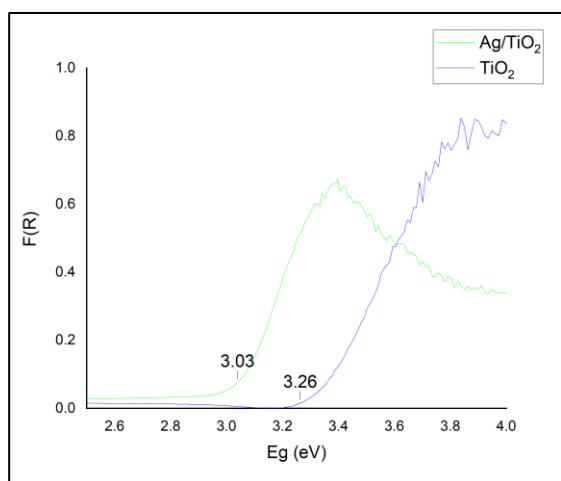


**Fig 6.** EDS analysis of Ag/TiO<sub>2</sub>

Photophysical properties of Ag/TiO<sub>2</sub> were characterized using UV – Vis DRS. **Fig. 7** shown that the presence of Ag caused the decrease reflectance of TiO<sub>2</sub> nanoparticle. Decreasing of the reflectance value (% R) was observed in the UV and visible light region, which is around 350 – 800 nm. TiO<sub>2</sub> strongly absorbs light in 400 nm, while Ag/TiO<sub>2</sub> absorption occurs in the visible region and edge redshift at a wavelength of 450 nm. A broadened absorption of Ag/TiO<sub>2</sub> was formed due to the SPR effect of AgNPs. Band gap of TiO<sub>2</sub> and Ag/TiO<sub>2</sub> calculated by Kubelka – Munk and Tauc equation. Tauc equation of TiO<sub>2</sub> and Ag/TiO<sub>2</sub> shown in **Fig. 8**. Band gap of TiO<sub>2</sub> and Ag/TiO<sub>2</sub> is 3,26 eV and 3,02 eV, respectively.



**Figure 7.** UV-Vis DRS analysis of TiO<sub>2</sub> and Ag/TiO<sub>2</sub>



**Figure 8.** Band gap of TiO<sub>2</sub> and Ag/TiO<sub>2</sub> by Tauc Plot equation

#### 4. CONCLUSION

We have successfully synthesized Ag/TiO<sub>2</sub> NPs from *Mirabilis jalapa* plant extract, as an eco-friendly, cost-effective, and non-toxic method. Ag/TiO<sub>2</sub> NPs were synthesized with the addition of *Mirabilis jalapa* plant extract as a reducing and stabilizing agent. The average size of the Ag/TiO<sub>2</sub> nanoparticles was  $\pm 160$  nm.

#### ACKNOWLEDGEMENT

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